

Determining Reactor Flux from Xenon-136 and Cesium-135 in Spent Fuel

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The ability to infer the reactor flux from spent fuel or seized fissile material would enhance the tools of nuclear forensics and nuclear nonproliferation significantly. We show that reactor flux can be inferred from the ratios of xenon-136 to xenon-134 and cesium-135 to cesium-137. If the average flux of a reactor is known, the flux inferred from measurements of spent fuel could help determine whether that spent fuel was loaded as a blanket or close to the mid-plane of the reactor. The cesium ratio also provides information on reactor shutdowns during the irradiation of fuel, which could prove valuable for identifying the reactor in question through comparisons with satellite reactor heat monitoring data. We derive analytic expressions for these correlations and compare them to experimental data and to detailed reactor burn simulations. The enrichment of the original uranium fuel affects the correlations by up to 3 percent, but only at high flux.

I. INTRODUCTION

Verification of reactor operations is an important component of nuclear nonproliferation and safeguards. Of particular importance is the neutron flux to which fuel was exposed during reactor operation, which might not necessarily be the same as the average flux quoted for a reactor. For example, if the spent fuel in question was loaded in the reactor as a blanket, it would generally see a lower flux than would the fuel loaded in the mid-plane. Thus, it is important to verify the flux from measurements of the spent fuel itself. Ratios of fission products in spent reactor fuel or released as fission gases during fuel reprocessing are used to deduce information about the irradiation history of the fuel. In the case of seized undetonated nuclear material, fission product ratios can enable inferences about the reactor used to generate the fuel, particularly when compared with reactor monitoring information. In this paper, we examine the correlation between the $^{136}\text{Xe}/^{134}\text{Xe}$ and $^{135}\text{Cs}/^{137}\text{Cs}$ fission product ratios and the thermal neutron flux used to irradiate the fuel. We also examine how the reactor power history, particularly reactor shutdowns, might also be extracted from these ratios. Correlations between a number of fission products and the reactor operations were observed experimentally by Maeck *et al.* [1]. In the present work we identify the underlying physics governing the dependence of the xenon and cesium ratios on reactor operation, we derive functional forms for these ratios, and we compare these with detailed reactor simulations and with the experimental data.

The sensitivity of the two fission product ratios to the reactor thermal neutron flux arises because of the competition between the 9.14 hour decay of xenon-135 to cesium-135 and the thermal neutron capture on xenon-135 to xenon-136, with an abnormally large cross section of 2.6×10^6 barns, Figure 1. For low flux, the majority of xenon-135 produced in fission β -decays to cesium-135, while for high flux most of the xenon-135 is transmuted to xenon-136. In contrast, reactor production of xenon-134 and cesium-137 is simply dependent on the number of fissions, because both of these nuclei are produced almost entirely as direct fission products or by the beta decay of other fission products.

The accuracy with which the cesium and xenon ratios can be measured depends on the size of the sample. Using resonance ionization mass spectrometry, Pibida *et al.* [2] have shown that the $^{135}\text{Cs}/^{137}\text{Cs}$ ratio can be determined reliably to 1% accuracy in very low-level samples containing large natural cesium backgrounds. This cesium ratio has been used [3] as a forensic tool for identifying the source of radioactive contamination in soil samples, though there is some evidence [4] to suggest that isotope fractionation may affect the cesium ratio in reactor effluences, depending on the location of the measured sample. Resonance ionization mass spectrometry has been shown [5] to be capable of determining ratios of naturally occurring xenon isotopes to better than 0.5% in samples of 10^{-12} cm^3 STP of xenon. For large samples, mass spectrometry of both xenon and cesium ratios can achieve an accuracy better than $1 : 10^4$.

The concentration of xenon-135 under different reactor conditions is well studied [6] because of the role it

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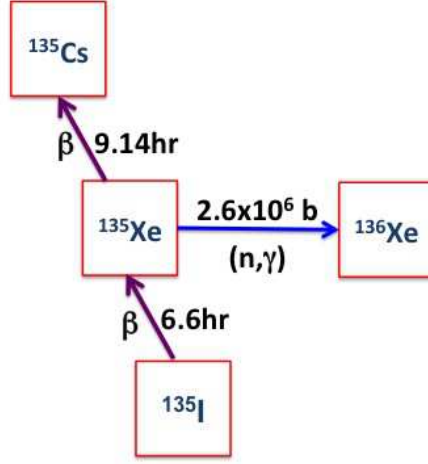


FIG. 1: The competition between the decay of xenon-135 to cesium-135 and the thermal neutron capture on xenon-135 to xenon-136 causes the relative concentrations of xenon-136 and cesium-135 in reactor fuel to be sensitive indicators of the thermal neutron flux.

plays as a serious reactor poison. If the reactor is operating in a steady state with a constant neutron flux, the concentration of xenon-135 reaches an equilibrium value that depends on the concentration of iodine-135 and on the rate of capture to xenon-136. Changes in the thermal flux cause short-term fluctuations in the level of xenon-135, and after about 40 to 50 hours of steady flux, the xenon-135 level settles to a new equilibrium value that reflects the new flux. Such changes in flux, as well as shutdowns and restarts of the reactor, also affect the $^{136}\text{Xe}/^{134}\text{Xe}$ and $^{135}\text{Cs}/^{137}\text{Cs}$ ratios. In this paper we quantify these effects and show how measurements of these two ratios could provide important information on reactor flux and power history.

II. THE DEPENDENCE OF ^{136}Xe ON THE THERMAL FLUX

Xenon-136 is produced by three main mechanisms in a reactor: as a direct fission product, from the β -decay of the fission fragment iodine-136, and from neutron capture on xenon-135. The lifetimes of the nuclei in the beta-decay chain leading to xenon-136 are short, and the first two mechanisms can be co-added and described in terms of the so-called cumulative fission yield for xenon-136. The cumulative fission yields for all fission products have been evaluated by England and Rider [7] and Wahl [8]; the cumulative fission yield for xenon-136 is ~ 7 percent (%) per fission for the fissioning isotopes of uranium and plutonium.

Under steady state conditions, the growth rate of xenon-136 in the reactor is,

$$\dot{N}_{136\text{Xe}} = \bar{f}_{136\text{Xe}}\Gamma_F + N_{135\text{Xe}}^{\text{equil}}\phi_T\sigma_a \quad (1)$$

$$= \bar{f}_{136\text{Xe}}\Gamma_F \left[1 + \left(\frac{\bar{f}_{135\text{Xe}}}{\bar{f}_{136\text{Xe}}} \right) \frac{\phi_T\sigma_a}{\lambda_{135\text{I}} + \phi_T\sigma_a} \right], \quad (2)$$

where Γ_F is the fission rate, \bar{f}_A is the burn-weighted cumulative fission yield of nucleus A , ϕ_T is the thermal

[1] The cumulative yields f_A are actinide dependent. Throughout this paper we use the notation \bar{f}_A to mean the cumulative yield for fragment A when weighted by the linear combination of burning actinides. Since the linear combination of actinides contributing to the burn changes with time, in general, the cumulative yields \bar{f}_A are also time dependent.

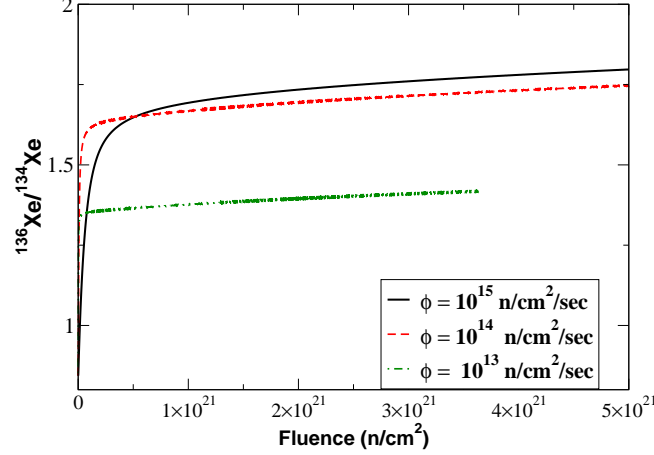


FIG. 2: The $^{136}\text{Xe}/^{134}\text{Xe}$ ratio equilibrates to a value that is determined by the thermal neutron flux, Equation (4). Thus, this ratio can be used to deduce the flux. The residual slope of the plateaus in the curves is caused by the slow evolution of the fuel composition, which is not explicitly taken account in Equation (4), except through the implied evolution of the cumulative yields \bar{f}_A ; see the discussion for Figure 6.

neutron flux, σ_a the thermal capture cross section on xenon-135, and $N_{^{135}\text{Xe}}^{equil}$ is the equilibrium value of xenon-135.

The dependence of the xenon-136 production rate on the magnitude of the thermal neutron flux falls between two limits for all thermal reactors,

$$\begin{aligned} \phi_T \sigma_a \gg \lambda_{135} : \dot{N}_{136} &= \bar{f}_{136} \Gamma_F \left(1 + \frac{\bar{f}_{135}}{\bar{f}_{136}} \right) \\ \phi_T \sigma_a \ll \lambda_{135} : \dot{N}_{136} &= \bar{f}_{136} \Gamma_F \end{aligned} \quad (3)$$

The production rate of xenon-134 only depends on the fission rate $\dot{N}_{134} = \bar{f}_{134} \Gamma_F$. Thus,

$$\frac{N_{136}}{N_{134}} = \frac{\bar{f}_{136} X_e}{\bar{f}_{134} X_e} \left(1 + \left(\frac{\bar{f}_{135} X_e}{\bar{f}_{136} X_e} \right) \frac{\phi_T \sigma_a}{\lambda_{135} + \phi_T \sigma_a} \right). \quad (4)$$

From Equation (4), the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio equilibrates on a (flux dependent) time scale of weeks. Figure 2 displays the situation for different values of the thermal flux.

A. Power Fluctuations and the Average Flux from $^{136}\text{Xe}/^{134}\text{Xe}$

If the reactor power is varied through variations in the thermal flux, the concentration of xenon-135 adjusts and reaches a new level. The value of the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio also reflects these variations. For this reason, previous studies [9–12] of xenon gases released during fuel reprocessing, for the purposes of extracting reactor operation information, omitted xenon-136 in their analyses. The concern was that, without knowledge of the power history, xenon-136 would be difficult to interpret. However, the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio is a direct measure of the *average flux* over the burn of the fuel. To show this we ran a series of simulations in which we varied the flux over different time periods. Figure 3 shows the effect on the concentration of xenon-135 of varying the power (1) on a daily basis and (2) every 15 days. In these calculations we kept the thermal neutron flux fixed at $\phi_T = 10^{13} \text{ n/cm}^2/\text{sec}$ for the first 12 hours each day (15 days of each month) and dropped it to

[2] If the fuel is irradiate for a long time ~ 1 year, production of all most isotopes by neutron capture becomes non-negligible. In the present work we are assuming that the burn times of interest are low enough to ignore neutron capture, except in the case of capture on xenon-135.

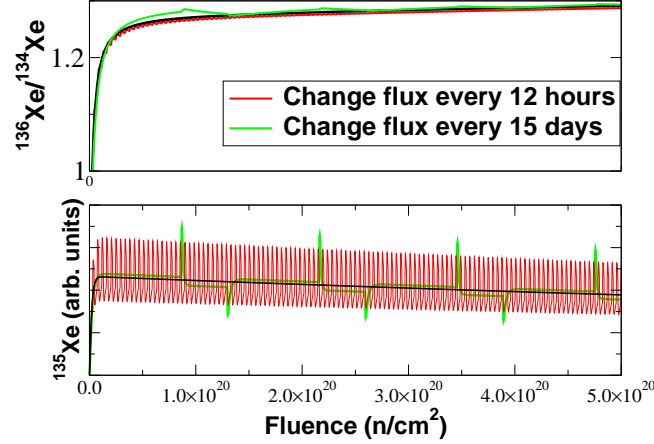


FIG. 3: Changes in the concentration of xenon-135 and in the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio with fluctuations in the thermal power. The red line corresponds to a change in the thermal flux by a factor of two every 12 hours, and the green line to change in the flux by a factor of two every 15 days. In both cases, the flux was varied between $5 \times 10^{12} \text{ n/cm}^2/\text{sec}$ and $10^{13} \text{ n/cm}^2/\text{sec}$. The black line corresponds to a constant flux of $7.5 \times 10^{12} \text{ n/cm}^2/\text{sec}$, which compares well with the results obtained with the fluctuating flux. Thus, with variations in the flux, the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio determines the average flux, which for most applications is the physical flux of interest.

$\phi_T = 0.5 \times 10^{13} \text{ n/cm}^2/\text{sec}$ for the second 12 hours (15 days). This caused the xenon-135 concentration to vary up and down, as displayed by the red (green) oscillating lines in the lower panel of Figure 3. The black line shows the effect of keeping the flux fixed at the average of these fluctuations, $\phi_T = 7.5 \times 10^{12} \text{ n/cm}^2/\text{sec}$. The upper panel of Figure 3 shows the resulting $^{136}\text{Xe}/^{134}\text{Xe}$ ratio, where it is clear that, in the presence of power variations, the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio determines the average thermal flux. This average flux is the physically meaningful quantity in determining flux-dependent physical properties of the spent fuel. At very high flux values, the nonlinearity in Equation (4) begins to skew the relation to the average flux, but this systematic effect does not become important until the flux is significantly higher than $10^{14} \text{ n/cm}^2/\text{sec}$.

III. THE RELATIONSHIP BETWEEN THERMAL FLUX AND THE $^{135}\text{Cs}/^{137}\text{Cs}$ RATIO

In this section we derive an analytic expression relating the concentration of cesium-135 to the thermal neutron flux. As seen in Figure 1, cesium-135 is produced through the decay chain $^{135}\text{I} \rightarrow ^{135}\text{Xe} \rightarrow ^{135}\text{Cs}$, which is complicated by the transmutation of xenon-135 via neutron capture. The number of cesium-135 atoms produced in a time t is given by

$$N_{135\text{Cs}}(t) = \lambda_{135\text{Xe}} \int_0^t dt' \left[\frac{\frac{\bar{f}_{135\text{I}}}{\bar{f}_{137\text{Cs}}} \dot{N}_{137\text{Cs}} - \dot{N}_{135\text{I}} - \dot{N}_{135\text{Xe}}}{\lambda_{135\text{Xe}} + \phi_T \sigma_a} \right]. \quad (5)$$

Here \dot{N}_A is the growth rate of nuclide A.

To solve for the $^{135}\text{Cs}/^{137}\text{Cs}$ ratio exactly requires numerical reactor burn simulations, which we present below. However, an analytic expression for this ratio can be obtained by assuming that, while the reactor is operating, the concentration of iodine-135 and xenon-135 are at their equilibrium values. We also assume that the measurements of cesium-135 are made after all of the iodine-135 and xenon-135 in the fuel has decayed. Thus, if the reactor is run with constant flux we can write

$$N_{135\text{Cs}} = \frac{\lambda_{135\text{Xe}}}{\lambda_{135\text{Xe}} + \phi_T \sigma_a} \frac{\bar{f}_{135\text{I}}}{\bar{f}_{137\text{Cs}}} N_{137\text{Cs}} + \frac{\phi_T \sigma_a}{\lambda_{135\text{Xe}} + \phi_T \sigma_a} \left(N_{135\text{I}}^{\text{equil}} + N_{135\text{Xe}}^{\text{equil}} \right) \quad (6)$$

$$= N_{137\text{Cs}} \frac{\bar{f}_{135\text{I}}}{\bar{f}_{137\text{Cs}}} \left[\frac{\lambda_{135\text{Xe}}}{\lambda_{135\text{Xe}} + \phi_T \sigma_a} + \frac{\phi_T \sigma_a}{\lambda_{135\text{Xe}} + \phi_T \sigma_a} \left(\frac{1}{\lambda_{135\text{I}} T_{\text{irrad}}} \right) \left(1 + \frac{\lambda_{135\text{I}}}{\lambda_{135\text{Xe}} + \phi_T \sigma_a} \right) \right]. \quad (7)$$

Here λ_{135I} is the decay constant for iodine-135, and T_{irrad} is the total irradiation time in the reactor, which we introduced by making the substitution, $N_{137Cs} = \bar{f}_{137Cs} \Gamma_F T_{irrad}$. The form of the second term, which gives rise to the term inversely proportional to the total irradiation time, arises from the proper treatment of startup transients in integrating Equation (5).

A. Effect of Reactor Shutdowns on the $^{135}\text{Cs}/^{137}\text{Cs}$ Ratio

We next consider the scenario in which the reactor is shutdown once or several times and restarted. Because xenon-135 is a serious reactor poison, shutdown times normally have to be long enough for all of the xenon-135 (and iodine-135) to decay. Thus, each time the reactor is shutdown the cesium-135 concentration is increased by the $^{135}\text{I} \rightarrow ^{135}\text{Xe} \rightarrow ^{135}\text{Cs}$ decay chain. Assuming that the flux is constant during operating periods, the effect of shutdowns is simply to add terms identical to the second term in Equation (7), one for each time the reactor is shutdown and restarted. If P represents the number of times the reactor is run and shutoff before the fuel is removed, with $P_{minimum} = 1$, the $^{135}\text{Cs}/^{137}\text{Cs}$ ratio is given by,

$$N_{135Cs}/N_{137Cs} = \frac{\bar{f}_{135I}}{\bar{f}_{137Cs}} \left[\frac{\lambda_{135Xe}}{\lambda_{135Xe} + \phi_T \sigma_a} + \frac{\phi_T \sigma_a}{\lambda_{135Xe} + \phi_T \sigma_a} \left(\frac{P}{\lambda_{135I} T_{irrad}^{total}} \right) \left(1 + \frac{\lambda_{135I}}{\lambda_{135Xe} + \phi_T \sigma_a} \right) \right]. \quad (8)$$

Here T_{irrad}^{total} is the sum of all the irradiation times to which the fuel was exposed. Comparing the magnitude of the main contribution to that of the correction to the $^{135}\text{Cs}/^{137}\text{Cs}$ ratio, it can be seen that the correction becomes significant for high flux. Thus, a measurement of the $^{135}\text{Cs}/^{137}\text{Cs}$ ratio in spent fuel could provide information on both the thermal flux and the burn history. The sensitivity to burn history is expressed in the ratio of the number of times the reactor was shutdown to the the total irradiation time scaled by the mean lifetime of ^{135}I . This dimensionless factor encapsulates the total dependence of the observed ratio on history.

B. Effect of Reactor Shutdowns on the $^{136}\text{Xe}/^{134}\text{Xe}$ Ratio

The correction to the $^{135}\text{Cs}/^{137}\text{Cs}$ ratio arising from reactor shutdowns, given by the second term in Equation (8), implies an equivalent correction to the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio of Equation (4).

$$\frac{N_{136Xe}}{N_{134Xe}} = \frac{\bar{f}_{136Xe}}{\bar{f}_{134Xe}} \left[1 + \frac{\phi_T \sigma_a}{\lambda_{135Xe} + \phi_T \sigma_a} \left(\frac{\bar{f}_{135Xe}}{\bar{f}_{136Xe}} \right) \left(1 - \frac{P}{\lambda_{135I} T_{irrad}^{total}} \left(1 + \frac{\lambda_{135I}}{\lambda_{135Xe} + \phi_T \sigma_a} \right) \right) \right]. \quad (9)$$

As mentioned above, the terms inversely proportional to the total irradiation time arise from proper treatment of startup transients. As can be seen from Equation (9) and Figure 5, the reactor shutdown correction to the flux dependence for the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio is small; this follows because of the way it appears in comparison to the constant 1 in the second term, a qualitatively different form compared to the expression in Equation (8).

IV. COMPARISON OF THE XENON AND CESIUM RATIOS TO EXPERIMENT

Maeck *et al.* measured [1] a series of fission isotope product ratios by irradiating highly enriched uranium targets in the Advanced Test Reactor (ATR) and in the Engineering Test Reactor (ETR) at Idaho National Laboratory. Each target was exposed to a different reactor thermal flux by placing the targets at different distances from the reactor mid-plane. The flux range was $6 \times 10^{12} - 1.5 \times 10^{14} \text{ n/cm}^2/\text{sec}$. The targets loaded in the ETR were more that 99% enriched uranium-235. The targets were divided into two groups, one that was irradiated for 20 days and one that was irradiated for 180 days over a 320 day period. The targets in the ATR were 93% enriched and were irradiated for approximately 100 days over a period of 335 days, with several reactor shutdowns over the 100 day irradiation period. The shape of the neutron flux for the two reactors is different, with the capture to fission ratio for uranium-235 being $\alpha_{235} \sim 0.18$ for the ETR and $\alpha_{235} \sim 0.215$ for the ATR. The flux shape also varied very slightly from target to target because of the difference in location within the reactor.

Figure 4 shows a comparison between the experimental data and the calculations for the $^{137}\text{Cs}/^{135}\text{Cs}$ isotopic ratio. (We note that, purely for display purposes, we follow Maeck *et al.* [1] and always graph the cesium ratio as $^{137}\text{Cs}/^{135}\text{Cs}$ and not the inverse.) The $^{137}\text{Cs}/^{135}\text{Cs}$ ratio scales approximately linearly with the flux.

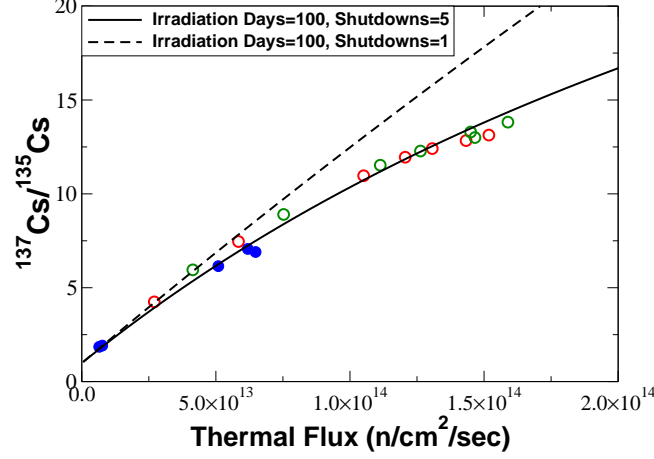


FIG. 4: The $^{137}\text{Cs}/^{135}\text{Cs}$ ratio. The measurements are the data of Maeck *et al.*[1]. The blue circles are data from 99% enriched uranium-235 samples that were irradiated in the ETR for either 20 days or 185 days (over a 320 day period). The green and red circles are data from 93% enriched samples that were irradiated in the ATR for 100 days over a period of 335 days. The dashed line is for a total irradiation time of 100 days, with one final shutdown. The straight line corresponds to an irradiation time of 100 days with a total of 5 reactor shutdowns. Both use the analytic expression (8). We note that this latter scenario leads to a $^{137}\text{Cs}/^{135}\text{Cs}$ ratio that is identical to that for 20 days irradiation, with one (final) shutdown, Equation (8). For high flux, it is clear that reactor shutdowns have a significant effect on the $^{137}\text{Cs}/^{135}\text{Cs}$ isotopic ratio.

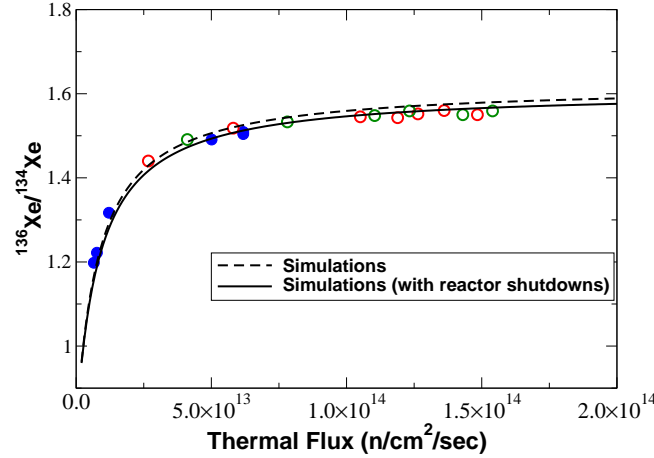


FIG. 5: The $^{136}\text{Xe}/^{134}\text{Xe}$ ratio. The measurements are the data of Maeck *et al.*[1]. The dashed line is for a total irradiation time of 100 days, with one final shutdown. The straight line corresponds to an irradiation time of 100 days with a total of 5 reactor shutdowns. We note that this latter scenario leads to a $^{136}\text{Xe}/^{134}\text{Xe}$ ratio that is identical to that for 20 days irradiation, with one (final) shutdown, Equation (8). As can be seen, reactor shutdowns have quite a small effect on the $^{136}\text{Xe}/^{134}\text{Xe}$ isotopic ratio. This is in contrast to the effect of shutdowns on the $^{137}\text{Cs}/^{135}\text{Cs}$ ratio.

Reactor shutdowns during the irradiation can have a significant effect on the the cesium ratio at high flux, $\phi_T \gtrsim 5 \times 10^{13} \text{ n/cm}^2/\text{sec}$.

Figure 5 shows the flux dependence of the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio. This ratio is also a sensitive function of the flux, though less sensitive than the cesium ratio. The $^{136}\text{Xe}/^{134}\text{Xe}$ varies by a factor of ~ 2 with the flux, Equation (4). It is most sensitive at low flux. Reactor shutdowns only change the xenon ratio slightly.

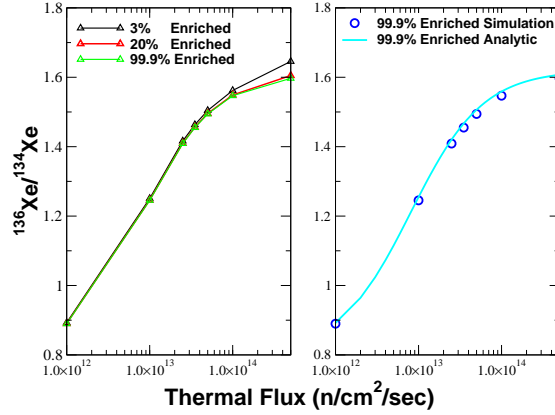


FIG. 6: Full reactor burn simulations for the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio. The left panel compares the full simulations for three uranium-235 enrichments. The right panel compares the results of the full reactor simulations with the analytic expression of Equation (9). As can be seen, the fuel enrichment does not affect the relation between the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio and the flux significantly, for flux values below about $5 \times 10^{13} \text{ n/cm}^2/\text{sec}$. For the highest flux considered, $5 \times 10^{14} \text{ n/cm}^2/\text{sec}$, the xenon ratio changed by 3% in going from 3% to 99.9% enrichment. This effect is due to differences in the plutonium growth rate and the corresponding changes in the cumulative fission yields (\bar{f}_A) appearing in Equation (9).

V. REACTOR BURN SIMULATIONS

The previous sections of this paper use our analytic expressions for the xenon and cesium isotopic ratios. It is important, however, to validate these results using full reactor simulations. For this we ran a series of simulations that included full production/depletion chains for all of the major actinides as well as the tellurium, iodine, xenon, and cesium fission fragment chains. In these simulations we examined three uranium fuel enrichments, 3%, 20%, and 99.9% uranium-235 enrichment. In all cases the total irradiation time was assumed to be 100 days, and we did not simulate shutdowns during the irradiation, since the shutdown corrections are the same for the analytic and simulated results. The results for the $^{136}\text{Xe}/^{134}\text{Xe}$ isotope ratio are shown in Figure 6, and those for the $^{137}\text{Cs}/^{135}\text{Cs}$ isotope ratio are shown in Figure 7. As can be seen, the analytic results provide a good representation of the full simulations. Both the xenon and cesium ratios are good indicators of the thermal flux, almost independent of the fuel enrichment. The fuel enrichment becomes important at high flux where a significant fraction of the fissions are from plutonium, for which the cumulative yields for xenon and cesium are different from those for uranium. But these effects change the xenon and cesium ratios by at most 3%.

VI. INFERRING INFORMATION ON REACTOR SHUTDOWNS

The correction to the cesium ratio from reactor shutdowns during the irradiation comes as a ratio of the total number of shutdowns to the total irradiation time. Therefore there is a degeneracy of possible solutions in relating the cesium ratio to the latter ratio. For example, a 20 day irradiation with one final shutdown would yield the same curve for flux versus cesium ratio as an irradiation of 100 days with 5 shutdowns. To break this degeneracy requires the use of an additional isotope ratio, and one straightforward possibility is to use a ratio that only depends on the thermal fluence or the number of fissions. An ideal probe of the thermal fluence is the ratio of $^{240}\text{Pu}/^{239}\text{Pu}$, since this ratio scales with the fluence but is not separately dependent on the flux or the enrichment of the in-going uranium fuel [13]. In Figure 8 we compare two scenarios, 75 days irradiation with one final shutdown and 150 days with two shutdowns. It is clear that, when plotted as a function of the plutonium ratio, these two scenarios become clearly distinguishable. If undetonated fissile material were to be seized, for example, these two ratios could be used to infer information on the reactor flux and the number of shutdowns.

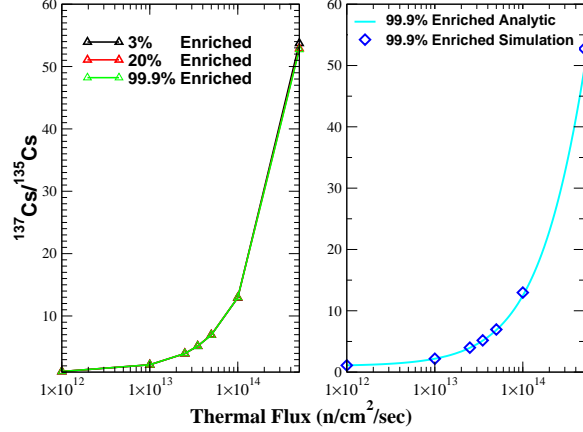


FIG. 7: Full reactor burn simulations for the $^{137}\text{Cs}/^{135}\text{Cs}$ ratio. The right panel compares the results of the full reactor simulations with the analytic expression of Equation (8). The left panel compares the full simulations for three uranium-235 enrichments. The fuel enrichment does not affect the relation between the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio and the flux significantly, except for the very highest flux values. At $\phi_T = 5 \times 10^{14} \text{ n/cm}^2/\text{sec}$, the cesium ratio changed by 2% in going from 3% to 99.9% enrichment.

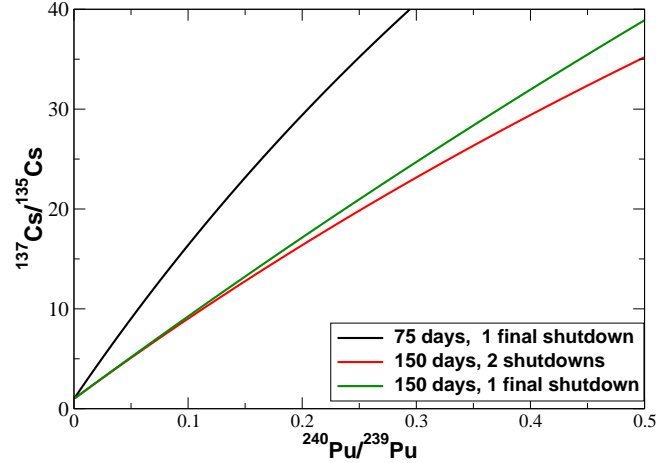


FIG. 8: The $^{137}\text{Cs}/^{135}\text{Cs}$ ratio as a function of the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio clear breaks the degeneracy between scenarios that involve the same ratio of number of reactor shutdowns to total irradiation time ($P/\lambda_{135I}T_{\text{irrad}}^{\text{total}}$). Thus, a measurement of both ratios in spent reactor fuel would make it possible to deduce the reactor flux and the number of times the reactor was shutoff over the time of the fuel irradiation. We note however, that for low flux there is little sensitivity to reactor shutdowns, and the cesium ratio directly determines the flux to which the fuel was exposed.

VII. DILUTION OF THE XENON RATIO INTO THE NATURAL ATMOSPHERE

In situations where the xenon ratio is measured as an effluent from fuel reprocessing at a finite distance from the stacks of the reprocessing facility, the dilution and mixing of the released stable xenon isotopes into the natural background of the air has to be taken into account. In this section, we estimate the volume of air into which the fission gases can be diluted, while maintaining useful information on the reactor flux from the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio.

We consider the general case in which two gas samples X and Y are mixed to form a blend B , such that the blend contains N_x and N_y atoms from samples X and Y , respectively, and $B = N_x/N_y$. If the ratio of any two stable xenon isotopes i, j in the original samples X and Y is R_{ijX} and R_{ijY} , respectively, then the ratio

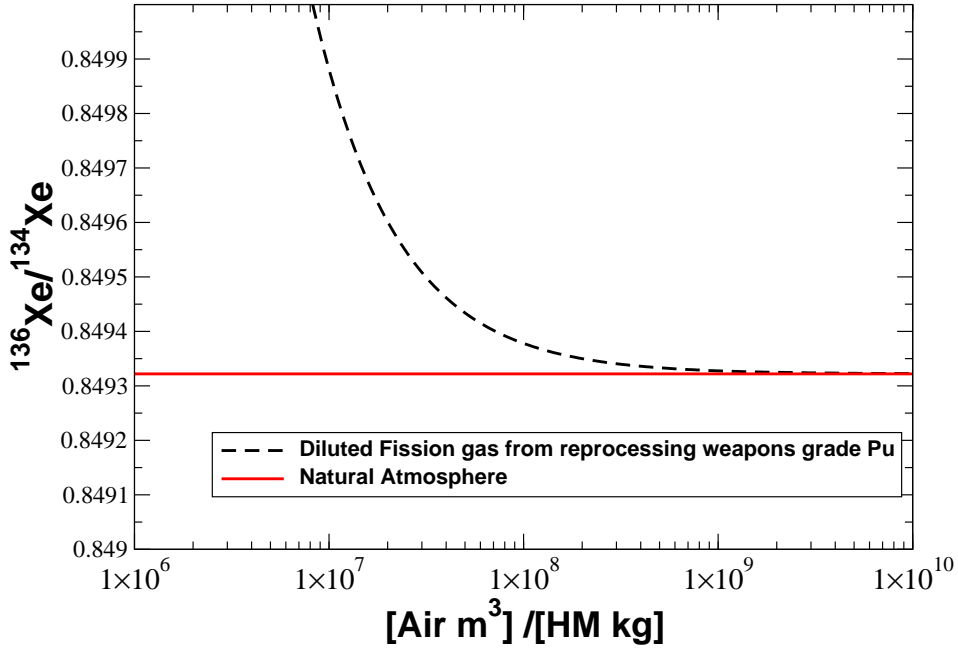


FIG. 9: The dilution of the fission produced $^{136}\text{Xe}/^{134}\text{Xe}$ ratio from mixing with the natural xenon in the atmosphere. The x-axis is the volume of air into which the fission gases are diluted per kilogram of heavy metal (HM) being reprocessed. Mass spectroscopic measurements of xenon ratios are accurate to better than 1 part in 10^4 . Thus, the $^{136}\text{Xe}/^{134}\text{Xe}$ ratio retains reactor relevant information for dilutions into up to $10^9 m^3$ of air.

in the blend R_{ijB} is given by the dilution equation [14, 15]

$$R_{ijB} = \frac{B \cdot R_{ijX} \sum_i R_{ijY} + R_{ijY} \sum_i R_{ijX}}{\sum_i R_{ijX} + B \cdot \sum_i R_{ijY}} \quad (10)$$

The number of atoms of xenon gas released during reprocessing depends on both the mass of heavy metal reprocessed and on the total reactor irradiation time to which the reprocessed fuel was exposed; for a given neutron flux, the irradiation time determines the grams of xenon per kilogram of heavy metal in the reprocessed fuel. The irradiation time can vary considerably, depending on the application. For example, irradiation times for medical isotopes are typically of the order of a few days, while those of weapons (reactor) grade plutonium are months (years). To estimate the effect of dilution on the xenon ratio, we ran reactor simulations for a burn history typical of what might be used for the production of weapons grade plutonium. We took the reactor flux to be $\phi = 10^{14} \text{ n/cm}^2/\text{sec}$ and the ^{235}U enrichment of the in-going uranium to be 3%. The irradiation time was assumed to be $5 \times 10^6 \text{ secs}$, corresponding to a total fluence of $5 \times 10^{20} \text{ n/cm}^2$ and a $^{240}\text{Pu}/^{239}\text{Pu}$ ratio of $\sim 7\%$.

Under these assumptions, the produced $^{136}\text{Xe}/^{134}\text{Xe}$ ratio was 1.5799. The effect of dilution of the fission gases into the atmosphere on this ratio, as a function of the volume of air, is shown in Figs. 9. As can be seen, the xenon ratio retains reactor information for dilutions up to about $10^9 m^3$ of air. A more detailed analysis of the dilution problem for other reprocessing scenarios, as well as the important issue of determining the blend B , are the subject of a second paper [16].

VIII. CONCLUSIONS

The $^{137}\text{Cs}/^{135}\text{Cs}$ and the $^{136}\text{Xe}/^{134}\text{Xe}$ isotope ratios in spent reactor fuel provide direct information on the reactor flux to which the fuel was exposed. The cesium ratio shows the greater sensitivity to the flux and varies by a factor of 50 over the thermal flux range $10^{12} - 5 \times 10^{14} \text{ n/cm}^2/\text{sec}$. At the lowest flux values ($10^{12} \text{ n/cm}^2/\text{sec}$), the cesium ratio is close to unity and approximately independent of the flux. As the flux increases the relative concentration of cesium-135 is reduced by the transmutation of xenon-135 to xenon-136 through rapid neutron capture. If the age of the fuel is known, allowing corrections for the 30 year half-life decay of cesium-137, the $^{137}\text{Cs}/^{135}\text{Cs}$ ratio is always greater than unity. A decay-corrected observed ratio above

unity is indicative of a reactor flux greater than $10^{12}n/cm^2/sec$. For large flux, the cesium ratio is significantly altered by reactor shutdowns during the irradiation of the fuel. If the spent fuel can be analyzed in detail, the number of shutdowns can be inferred by comparing the $^{137}Cs/^{135}Cs$ ratio to the $^{240}Pu/^{239}Pu$ ratio.

The $^{136}Xe/^{134}Xe$ ratio varies by a factor of about 2 over the flux range $10^{12} - 5 \times 10^{14}n/cm^2/sec$. If the flux and power of the reactor are varied over the burn history, the xenon ratio determines the *average* flux to which the fuel was exposed. Thus, if xenon gases released in fuel reprocessing are measured, the $^{136}Xe/^{134}Xe$ ratio determines the physically relevant flux for reactor burn history analyses. Finally, we note that the xenon ratio shows most sensitivity to the flux for the lower flux values, whereas the cesium ratio is more sensitive in the higher flux range.

IX. ACKNOWLEDGEMENT

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